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IN RE APPLICATION OF

NOBUO KAIFU, ET AL. : EXAMINER: HEINCER, L. J.

SERIAL NO: 10/574,774

FILED: MAY 1, 2007 : GROUP ART UNIT: 1796

RCE FILED: FEBRUARY 17, 2009

FOR: PROCESS FOR PRODUCING RESORCINOL-FORMALIN RESIN

APPEAL BRIEF

COMMISSIONER FOR PATENTS ALEXANDRIA, VIRGINIA 22313

SIR:

This is an appeal of the Rejection dated June 11, 2009 of twice-rejected Claims 1-4, 6-18 and 20-23. A Notice of Appeal is **submitted herewith**.

I. REAL PARTY IN INTEREST

The real party in interest in this appeal is Hodogaya Chemical Co., Ltd, having an address at 66-2, Horikawa-cho, Saiwai-ku, Kawasaki-shi, Kanagawa, Japan 212-8588.

II. RELATED APPEALS AND INTERFERENCES

Appellants, Appellants' legal representative and the assignee are aware of no appeals, interferences, or judicial proceedings which may be related to, directly affect or be directly affected by or have a bearing on the Board's decision in this appeal.

III. STATUS OF THE CLAIMS

Claims 1-4, 6-18 and 20-23 stand rejected and are herein appealed. Claims 5 and 19 have been canceled.

IV. STATUS OF THE AMENDMENTS

No amendment under 37 CFR 1.116 has been filed.

V. SUMMARY OF THE CLAIMED SUBJECT MATTER

A summary of the claimed subject matter on appeal, as claimed in independent Claims 1 and 15, is mapped out below, with reference to page and line numbers in the specification added in [bold] after each element.

- Claim 1. A process for producing a resorcinol-formalin resin containing no inorganic salts, which comprises: [page 6, lines 12-14]
- (1) adding, to a water solvent, resorcinol (A) in an amount of 20 to 150 parts by weight relative to 100 parts by weight of water, an inorganic salt (B) in an amount of 20 to 80 parts by weight relative to 100 parts by weight of water, and an organic solvent (C) having a solubility parameter of 7.0 to 12.5 and capable of dissolving the resorcinol-formalin resin in an amount of 10 to 200 parts by weight relative to 100 parts by weight of resorcinol (A), thereby forming a mixture; [page 6, line 20 to page 7, line 3]
- (2) stirring the mixture at a liquid temperature not higher than the boiling point of the organic solvent (C) to give a two-phase system containing no remaining solid matter; [page 7, lines 4-7]
- (3) adding a catalytic amount of an organic acid or inorganic acid (D) to the mixture formed in step (2); [page 7, lines 8-9]

- (4) adding 1 to 40% formalin (E) dropwise to the mixture formed in step (3) in a molar ratio of formaldehyde/resorcinol of 0.3 to 0.8 under stirring over a period of 1 to 300 minutes thereby forming a reaction system, while maintaining the reaction system at 0 to 60°C; [page 7, lines 10-13]
- (5) stirring the mixture formed in step (4) for further 10 to 60 minutes after the completion of the dropwise addition to cause a liquid-liquid heterogeneous reaction to proceed; [page 7, lines 14-17]
- (6) allowing the reaction system to stand while maintaining it at the temperature of the reaction to separate it into two layers, which are an aqueous layer and a reaction product layer; [page 7, lines 18-20]
 - (7) removing the aqueous layer; [page 7, line 21]
- (8) adding an organic solvent (C) in an amount of 1 to 5 equivalents to the amount of the reaction product to the reaction product layer which is an organic solvent layer to effect dilution; [page 7, lines 22-25]
- (9) adding water to the reaction product layer in an amount which is half of the amount of the organic solvent; [page 7, line 26 to page 8, line 2]
- (10) stirring the reaction system after adding of said organic solvent (C) and said water while maintaining its temperature to be not higher than the boiling point; [page 8, lines 3-4]
- (11) separating the reaction system of step (10) into two layers, including an aqueous layer, after allowing the reaction system to stand; and then [page 8, lines 5-6]
- (12) removing the aqueous layer to obtain a resorcinol-formalin resin, [page 8, lines 7-8]

wherein said reaction is a one-stage reaction and liquid-liquid distribution is conducted in the same reactor, [page 6, lines 17-19] and

wherein the organic solvent (C) is an organic solvent represented by the following general formula [1]:

General formula [1]:

[page 10, lines 1-5th line from bottom]

wherein m represents 0 or 1, n represents 0 or 1, and R₁, R₂, R₃, R₄, and R₅ each independently represents a methyl group, an ethyl group, an n-propyl group, a 2-propyl group, an n-butyl group, a secondary butyl group, an isobutyl group, or a tertiary butyl group; R₁ and R₂ may be combined to form a ring when m=n=0, R₂ and R₃ may be combined to form a ring when m=1 and n=0, and R₃ and R₄ may be combined to form a ring when m=n=1, with the proviso that when X is -O- and m=n=0, R₁ and R₂ are combined to form a ring.

Claim 15. A resorcinol-formalin resin containing no inorganic salts, [page 14, lines 20-21] produced by a production process comprising: [page 14, line 24]

(1) adding, to a water solvent, resorcinol (A) in an amount of 20 to 150 parts by weight relative to 100 parts by weight of water, an inorganic salt (B) in an amount of 20 to 80 parts by weight relative to 100 parts by weight of water, and an organic solvent (C) having a solubility parameter of 7.0 to 12.5 and capable of dissolving the resorcinol-formalin resin in

an amount of 10 to 200 parts by weight relative to 100 parts by weight of resorcinol (A),thereby forming a mixture; [page 15, lines 3-11]

- (2) stirring the mixture at a liquid temperature not higher than the boiling point of the organic solvent (C) to give a two-phase system containing no remaining solid matter; [page 15, lines 12-14]
- (3) adding a catalytic amount of an organic acid or inorganic acid (D) to the mixture formed in step (2); [page 15, lines 16-17]
- (4) adding 1 to 40% formalin (E) dropwise to the mixture formed in step (3) in a molar ratio of formaldehyde/resorcinol of 0.3 to 0.8 under stirring over a period of 1 to 300 minutes thereby forming a reaction system, while maintaining the reaction system at 0 to 60°C; [page 15, lines 18-21]
- (5) stirring the mixture formed in step (4) for further 10 to 60 minutes after the completion of the dropwise addition to cause a liquid-liquid heterogeneous reaction to proceed; [page 15, lines 22-24]
- (6) allowing the reaction system to stand while maintaining it at the temperature of the reaction to separate it into two layers, which are an aqueous layer and a reaction product layer; [page 15, line 25 to page 16, line 2]
 - (7) removing the aqueous layer; [page 16, line 3]
- (8) adding an organic solvent (C) in an amount of 1 to 5 equivalents to the amount of the reaction product to the reaction product layer which is an organic solvent layer to effect dilution; [page 16, lines 4-7]
- (9) adding water to the reaction product layer in an amount which is half of the amount of the organic solvent; [page 16, lines 8-10]

Application No. 10/574,774 Appeal Brief

(10) stirring the reaction system after adding of said organic solvent (C) and said water while maintaining its temperature to be not higher than the boiling point; [page 16, lines 11-12]

- (11) separating the reaction system of step (10) into two layers, including an aqueous layer, after allowing the reaction system to stand; and then [page 16, lines 13-14]
- (12) removing the aqueous layer to obtain a resorcinol-formalin resin, [page 16, lines 15-16]

wherein said reaction is a one-stage reaction and liquid-liquid distribution is conducted in the same reactor, [page 14, line 25 to page 15, line 2] and

wherein the organic solvent (C) is a ketone represented by the following general formula [1]:

General formula [1]:

$$\left(R_3-Y-\right)_m$$
 $R_1-X-R_2-\left(-Z-R_4\right)_n$

wherein m represents 0 or 1, n represents 0 or 1, and R₁, R₂, R₃, R₄, and R₅ each independently represents a methyl group, an ethyl group, an n-propyl group, a 2-propyl

group, an n-butyl group, a secondary butyl group, an isobutyl group, or a tertiary butyl group; R_1 and R_2 may be combined to form a ring when m=n=0, R_2 and R_3 may be combined to form a ring when m=1 and n=0, and R_3 and R_4 may be combined to form a ring when m=n=1. [page 18, lines 10 to page 19, line 5]

VI. GROUNDS OF REJECTION

Ground (A)

Claims 1-4 and 6-12 stand rejected under 35 U.S.C. § 103(a) as unpatentable over JP 2003-277308 (Honda) in view of US 3,814,713 (Honda et al) and US 4,376,854 (Yamaguchi et al) as evidenced by US 4,876,324 (Nakano et al).

Ground (B)

Claim 13 stands rejected under 35 U.S.C. § 103(a) as unpatentable over <u>Honda</u> in view of <u>Honda et al</u> and <u>Yamaguchi et al</u> as evidenced by <u>Nakano et al</u>, and further in view of US 5,368,928 (<u>Okamura et al</u>).

Ground (C)

Claim 14 stands rejected under 35 U.S.C. § 103(a) as unpatentable over <u>Honda</u> in view of <u>Honda et al</u> and <u>Yamaguchi et al</u> as evidenced by <u>Nakano et al</u>, and further in view of US 2004/0116647 (<u>Swedo</u>).

Ground (D)

Claims 15-18 and 20-23 stand rejected under 35 U.S.C. § 103(a) as unpatentable over Honda in view of Honda et al and Yamaguchi et al as evidenced by Nakano et al.

VII. ARGUMENT

Claims 1-4 and 6-12 stand rejected under 35 U.S.C. § 103(a) as unpatentable over Honda in view of Honda et al and Yamaguchi et al as evidenced by Nakano et al. That rejection is untenable and should not be sustained.

As recited in Claim 1, an embodiment of the present invention is a process for producing a resorcinol-formalin resin containing no inorganic salts, which comprises:

- (1) adding, to a water solvent, resorcinol (A) in an amount of 20 to 150 parts by weight relative to 100 parts by weight of water, an inorganic salt (B) in an amount of 20 to 80 parts by weight relative to 100 parts by weight of water, and an organic solvent (C) having a solubility parameter of 7.0 to 12.5 and capable of dissolving the resorcinol-formalin resin in an amount of 10 to 200 parts by weight relative to 100 parts by weight of resorcinol (A) thereby forming a mixture;
- (2) stirring the mixture at a liquid temperature not higher than the boiling point of the organic solvent (C) to give a two-phase system containing no remaining solid matter;
- (3) adding a catalytic amount of an organic acid or inorganic acid (D) to the mixture formed in step (2);
- (4) adding 1 to 40% formalin (E) dropwise to the mixture formed in step (3) in a molar ratio of formaldehyde/resorcinol of 0.3 to 0.8 under stirring over a period of 1 to 300 minutes thereby forming a reaction system, while maintaining the reaction system at 0 to 60°C;
- (5) stirring the mixture formed in step (4) for further 10 to 60 minutes after the completion of the dropwise addition to cause a liquid-liquid heterogeneous reaction to proceed;

Application No. 10/574,774 Appeal Brief

- (6) allowing the reaction system to stand while maintaining it at the temperature of the reaction to separate it into two layers, which are an aqueous layer and a reaction product layer;
 - (7) removing the aqueous layer;
- (8) adding an organic solvent (C) in an amount of 1 to 5 equivalents to the amount of the reaction product to the reaction product layer which is an organic solvent layer to effect dilution;
- (9) adding water to the reaction product layer in an amount which is half of the amount of the organic solvent;
- (10) stirring the reaction system after adding of said organic solvent (C) and said water while maintaining its temperature to be not higher than the boiling point;
- (11) separating the reaction system of step (10) into two layers, including an aqueous layer, after allowing the reaction system to stand; and then
- (12) removing the aqueous layer to obtain a resorcinol-formalin resin,
 wherein said reaction is a one-stage reaction and liquid-liquid distribution is
 conducted in the same reactor, and

wherein the organic solvent (C) is an organic solvent represented by the following general formula [1]:

General formula [1]:

wherein m represents 0 or 1, n represents 0 or 1, and R_1 , R_2 , R_3 , R_4 , and R_5 each independently represents a methyl group, an ethyl group, an n-propyl group, a 2-propyl group, an n-butyl group, a secondary butyl group, an isobutyl group, or a tertiary butyl group; R_1 and R_2 may be combined to form a ring when m=n=0, R_2 and R_3 may be combined to form a ring when m=1 and n=0, and R_3 and R_4 may be combined to form a ring when m=n=1, with the proviso that when X is -O- and m=n=0, R_1 and R_2 are combined to form a ring.

Another embodiment of the present invention, as recited in Claim 15, is, in effect, the product made by the process of Claim 1, except that the organic solvent (C) is limited to a ketone.

In sum, the present process invention is directed to a process comprising: adding, to a water solvent, resorcinol, an inorganic salt (molecular weight controlling agent), and a specific organic solvent (C) (having a solubility parameter of 7.0 to 12.5); stirring the resulting mixture to give a two-phase system; adding an acid catalyst; dropwise adding formalin thereto; and stirring the resulting mixture to cause a liquid-liquid heterogeneous reaction. The process of the invention provides a resorcinol-formalin resin containing no inorganic salts, having a moderate flowability, and having reduced contents of resorcinol monomer and resorcinol pentanuclear or higher nuclear bodies. The present product

invention is directed to a product made by the process, except that the organic solvent (C) is limited to a ketone. As discussed in further detail under Ground (D), the product claims are separately patentable.

As described in the specification under Background Art, beginning at page 1, 2nd paragraph, resorcinol-formalin resins are known in the art, but flowability has been problematical, as well as the suggested solutions thereof. An object of the present invention, as described in the specification at page 5, 1st full paragraph, is to provide a process for producing a resorcinol-formalin resin containing no salts and having a moderate flowability when transformed into an aqueous solution, which is capable of reducing both the content of resorcinol monomer and the content of resorcinol-formalin resin of resorcinol pentanuclear or higher nuclear bodies in a one-stage reaction. Applicants have been able to attain this objective by adopting a liquid-liquid heterogeneous reaction using a specific range of organic solvents, i.e., organic solvents having a solubility parameter within a particular range, as described in the specification at the paragraph bridging pages 5 and 6, and as discussed above.

A liquid-liquid heterogeneous reaction is one in which the reaction is carried out in a mixed solvent of water and a solvent. In the present invention, it is essential to use the organic solvent.

As described at the paragraph bridging pages 20 and 21 of the specification, the advantages derived from the adoption of the liquid-liquid heterogeneous reaction are as follows: (1) since the reaction proceeds faster in an aqueous phase and the existing ratio of resorcinol is in favor to the aqueous phase, the amount of unreacted resorcinol can be reduced; (2) the solubility of the resorcinol polynuclear bodies (especially, tetranuclear or higher nuclear bodies wherein three-dimensional structures are present) are rapidly decreased with the progress of the reaction and hence they migrate into an organic phase; furthermore,

since the reaction proceeds slower in the organic phase, the formation of resorcinol pentanuclear or higher nuclear bodies are inhibited; (3) since the reaction proceeds in both of the aqueous phase and the organic phase, the reaction time can be shortened; (4) the reaction is completed as an one-stage reaction between resorcinol and formalin and a step for removing the resorcinol polynuclear bodies after the reaction is not necessary, and (5) for the above reasons, the formation of both of unreacted resorcinol and resorcinol polynuclear bodies can be reduced, the reaction time can be shortened, and also the reaction can be completed in a one-stage reaction, so that the process is extremely advantageous in view of the cost.

The specification herein contains comparative data demonstrating the significance of various parameters of the presently-claimed process and product obtained thereby. Examples 1-11 are according to the presently-claimed invention. Comparative Examples 1 and 2, which exemplify a solid-liquid heterogeneous reaction, without an organic solvent, are for purposes of comparison. The results for Comparative Examples 1 and 2 were deficient, as described in the specification at page 37, lines 1-15, and page 38, lines 5-9, respectively.

The applied prior art neither discloses the present invention or its advantages, as above discussed.

Honda discloses a low-molecular weight resorcinol-formaldehyde reaction product having a small content of unreacted resorcinol and excellent flowability, wherein resorcinol and formaldehyde are reacted in the presence of an acidic catalyst, a molecular weight controlling agent [0009] is added and the reaction is carried out in water. The kind and amount of the molecular weight controlling agent, which may be a particular organic or inorganic salt [0013], are selected so that a large amount of dimer and trimer of resorcinol is precipitated. A precipitated product generated when the concentration of the generated

substance reaches the saturated concentration and no further dissolution in water is possible is collected.

For collecting the reaction product, it is good to squeeze out the mother liquid, for example, by squeezing under pressure, but this cannot completely remove the mother liquid. If necessary, the product is washed with a resorcinol washing liquid. Further, if necessary, a selected solvent method using a solvent that dissolves the reaction product but does not dissolve the molecular weight controlling agent is used. When the molecular weight controlling agent is calcium chloride, 2-butanone may be used for the selected solvent method [0021] and [0022].

Thus, according to <u>Honda</u>, the reaction of resorcinol and formaldehyde is carried out in an **aqueous solvent** in the presence of a molecular weight controlling agent, such as calcium chloride. The low-molecular weight resorcinol-formaldehyde reaction product is obtained as a precipitate generated when the concentration reaches the saturated concentration and no further dissolution becomes possible. In the present invention, on the other hand, a specific organic solvent having a solubility parameter falling within a specific range and immiscible with water, and a heterogeneous system reaction is carried out in the presence of an inorganic salt, such as calcium chloride, serving as a molecular weight controlling agent.

According to <u>Honda</u>, the reaction product precipitates after reaching the saturated concentration, because no further dissolution in water is possible. In the present invention, on the other hand, since an organic solvent, such as methyl ethyl ketone (MEK), having dissolution ability with respect to the reaction product as compared with water is used, precipitation does not occur and the reaction product migrates to the organic solvent side.

The resorcinol-formalin resin obtained by the present invention has a balanced distribution over the whole molecular weight region from low molecular weight (resorcinol

monomer) to high molecular weight (pentanuclear or higher nuclear bodies) as compared to the resin obtained by <u>Honda</u>, and further, has flowability. Furthermore, the objective low-molecular weight resorcinol-formalin resin is dissolved in the organic solvent, and does not contain an inorganic salt such as calcium chloride serving as the molecular weight controlling agent. For this reason, the product shows an excellent effect in adhesion with another material.

Moreover, the process of the invention does not require, as <u>Honda</u> requires, "squeezing under pressure", "washing with a resorcinol washing liquid" or "selected solvent method" in collecting the reaction product.

Honda et al discloses an adhesive composition for bonding rubber materials to any of various materials including synthetic and natural polymers, metals, silicate and ceramic materials with only a single application, which composition comprises a rubber latex, a high molecular weight resorcinol-formaldehyde resin and a low molecular weight resorcinolformaldehyde resin, wherein the latter component contains unreacted resorcinol in an amount less than 15 wt% (Abstract). The low molecular weight resorcinol-formaldehyde (RF) resin is disclosed as "must be prepared carefully according to a particular process" (sentence bridging columns 2 and 3). This component is obtained by reacting resorcinol with formaldehyde in a mole ratio of 1 to 0.05-0.8, preferably in water and without a catalyst or in the presence of a very small amount of an acidic catalyst (column 4, lines 6-12). Honda et al discloses further that in order to synthesize the low molecular weight RF resin involving very little or no amount of resorcinol polymers higher than the pentamer inclusive, it is necessary to react resorcinol with formaldehyde while limiting the amount of the latter to less than equimole of the former (column 4, lines 34-38). After completion of the reaction, the reaction product is dissolved in a ketone, and the extract is repeatedly washed with water or extracted therewith so that unreacted resorcinol is selectively transferred to the water phase,

thereby resulting in RF resin having unreacted resorcinol in an amount decreased down to a predetermined ratio, wherein methyl isobutyl ketone and diisobutyl ketone are the preferred ketones, which ketones are used in an amount of 4 to 6 times the lower molecular weight RF resin (column 5, lines 1-43).

Thus, <u>Honda et al</u> uses the ketone solvent in a post-treatment after completion of the reaction. By the repeated water extractions from the organic solvent layer in which the reaction product is dissolved, the content of the unreacted resorcinol remaining in the resorcinol-formaldehyde resin is reduced to less than 15 wt%.

As discussed above, the ketone solvent is used for the post-treatment after completion of the reaction. The reaction *per se* is carried out in an **aqueous system**. The reaction system is also different from that of the present invention. Further, the low-molecular weight resorcinol-formaldehyde resin and the high-molecular weight resorcinol-formaldehyde resin are prepared under separate conditions, and they are blended at a predetermined ratio to obtain an objective adhesive composition. The present invention, on the other hand, is directed to a production process in which all the steps including the one-stage reaction and the liquid-liquid distribution are carried out in the same single reactor. This provides a further distinction over Honda et al.

In addition, according to the process of the present invention, when using a ketone such as MEK as the organic solvent, the resin and the solvent react to form a structure in which the solvent is incorporated as part of the resin structure, hence providing a ketone-modified resorcinol-formalin resin (novel resin). For this reason, it provides a modified structure that is promising to dramatically increase the adhesion property as compared to conventional resorcinol-formalin resins. This contributes improvements in quality and performance. Such advantages are discussed further under Ground (D), *infra*.

Yamaguchi et al discloses preparing novolak-type copolymers by reacting an m-substituted phenol with formaldehyde in the presence of an alkaline catalyst to form a resoltype precondensate and a specific amount of unreacted formaldehyde, and then adding the resol-type precondensate and unreacted formaldehyde to a mixture of an acid and a resorcinol (paragraph bridging columns 1 and 2), wherein the resol-type precondensate can be prepared in an aqueous medium or in a water-organic solvent medium, wherein the organic solvent may be, *inter alia*, an aliphatic ketone. All of the examples in Yamaguchi et al use an aqueous medium.

Nakano et al discloses preparing o-cresol-novolak resins in a polar organic solvent in the presence of an acid catalyst, wherein the solvent is a C_{3-12} aliphatic alcohol, C_{3-6} glycol ether, benzyl alcohol or a C_{2-6} aliphatic carboxylic acid (paragraph bridging columns 4 and 5). Nakano et al discloses further that the presence of water should be, in effect, minimized because it results in a heterogeneous reaction system (column 6, lines 24-40).

The Examiner holds that it would have been obvious to have used the ketone of Yamaguchi et al in the process of Honda, and that the motivation to do so would have been, as suggested by Nakano et al, a biphasic reaction medium will lower the molecular weight of a phenol formaldehyde resin, relying on the disclosure in Nakano at column 6, lines 24-40.

In reply, neither <u>Yamaguchi et al</u> nor <u>Nakano et al</u> disclose or suggest the use of an organic solvent in the particular reaction disclosed by <u>Honda</u>, which involves reacting resorcinol and formaldehyde in the presence of an acidic catalyst. As discussed above, <u>Yamaguchi et al</u> optionally includes an organic solvent in their reaction medium to prepare a **resol-type** precondensate, which is obtained by reacting in an **alkaline catalyst**, not the acid catalyst of <u>Honda</u>, or the acid catalyst of the present invention. Moreover, <u>Nakano et al</u> actually teaches away from the use of a heterogeneous reaction, which results from the presence of water along with the polar organic solvent therein, as discussed above.

The Examiner acknowledges that <u>Honda</u> does not disclose diluting his product with an organic solvent, and thus relies on <u>Honda et al</u>. The Examiner holds that it would have been obvious to have used the solvent dilution and water extraction step of <u>Honda et al</u> in the process of <u>Honda</u>, and the motivation to do so would have been, as <u>Honda et al</u> suggests, to reduce the amount of unreacted resorcinol in the composition, relying on the disclosure in <u>Honda et al</u> at page 5, lines 36-40.

In reply, Applicants have discussed above the deficiencies in the combination of Honda and Honda et al, which are both drawn to aqueous systems.

The Examiner finds that <u>Honda</u> does not disclose adding his acid catalyst after the mixing/stirring of the other reaction components, but holds that the selection of any order of mixing ingredients is *prima facie* obvious. The Examiner then holds that it would have been obvious to have added the catalyst after mixing the other components, but prior to the addition of the formaldehyde.

In reply, since the presently-claimed invention is a liquid-liquid heterogeneous reaction, it is important to establish the two-phase system prior to adding the catalyst. Thus, in the present invention, one of ordinary skill in the art would appreciate that at least this particular order is required. One of ordinary skill in the art would not obtain such knowledge from <u>Honda</u>.

For all the above reasons, it is respectfully requested that this rejection be REVERSED.

Ground (B)

Claim 13 stands rejected under 35 U.S.C. § 103(a) as unpatentable over <u>Honda</u> in view of <u>Honda et al</u> and <u>Yamaguchi et al</u> as evidenced by <u>Nakano et al</u>, and further in view of <u>Okamura et al</u>. That rejection is untenable and should not be sustained.

Okamura et al is directed to a water-based treating liquid for a rubber-reinforcing glass fiber cord, which contains as a solid component a composition consisting essentially of:

- (1) a rubber latex
- (2) an alkylated chlorosulfonated polyethylene latex, and
- (3) a water-soluble resorcinol-formaldehyde condensate, wherein, based on the total amount of the components (1) and (2), the amount of the component (2) is 10 to 70% by weight and the amount of the component (3) is 5 to 20% by weight (column 3, lines 5-18).

The resorcinol-formaldehyde resin disclosed in <u>Okamura et al</u> is necessarily a resole-type resin obtained by using an alkali as a condensation agent, which is clearly distinct from a novolak-type resin obtained by using an acidic catalyst, such as hydrochloric acid, as a condensation agent. Also, the production process is different from that of the present invention. Okamura et al is, in essence, irrelevant.

In effect, Okamura et al does not remedy any of the deficiencies in the combination of the other prior art, as discussed under Ground (A), *supra*.

For all the above reasons, it is respectfully requested that this rejection be REVERSED.

Ground (C)

Claim 14 stands rejected under 35 U.S.C. § 103(a) as unpatentable over <u>Honda</u> in view of <u>Honda et al</u> and <u>Yamaguchi et al</u> as evidenced by <u>Nakano et al</u>, and further in view of <u>Swedo</u>. That rejection is untenable and should not be sustained.

Swedo relates to a particular phenol resin, and discloses various compounds to be used as a hardener and/or a catalyst for curing a phenol resin. Example 13 therein is drawn to a process for synthesizing a phenol-formalin resin (resole-type resin) in which a "sodium hydroxide solution" is used as a condensation agent. After the reaction in the aqueous

system, hydrochloric acid is added for neutralization, and then the upper, aqueous layer is decanted to obtain a resole layer. The resole is diluted with acetone, and the solution is dried over anhydrous magnesium sulfate, followed by filtration and removal of the acetone, to thereby obtain the resole-type resin. Thus, like <u>Okamura et al</u>, *supra*, <u>Swedo</u> is, in essence, irrelevant.

In effect, <u>Swedo</u> does not remedy any of the deficiencies in the combination of the other prior art, as discussed under Ground (A), *supra*.

For all the above reasons, it is respectfully requested that this rejection be REVERSED.

Ground (D)

Claims 15-18 and 20-23 stand rejected under 35 U.S.C. § 103(a) as unpatentable over Honda in view of Honda et al and Yamaguchi et al as evidenced by Nakano et al. That rejection is untenable and should not be sustained.

Everything stated above under Ground (A) is hereby incorporated by reference. In addition, the rejected claims under Ground (D) are also separately patentable from the rejected claims under Ground (A).

A feature of Claim 15 and claims dependent thereon resides in modification of the resorcinol-formalin resin by the ketone compound solvent. When a ketone solvent is used, the resorcinol-formalin resin is modified with the ketone. The ketone modification contributes significant enhancement of performance such as adhesion between a rubber tire and a polyester resin, as compared to the case of conventional resorcinol-formalin resins. The above is supported in the specification by various Examples. In Examples 1 to 5, using a ketone compound as an organic solvent, calculated yields are from 102 to 111% as

summarized in the Table below. A yield over 100% is evidence of the modification with the ketone compound.

On the other hand, tetrahydrofuran (THF) used in Example 6 has an ether structure and does not react with the resorcinol-formalin resin. Further, resins dissolved in tetrahydrofuran partly transfers to the aqueous layer. Hence, the yield is low.

Regarding Examples 7 to 11, the yields cannot be calculated because the amounts of the resins obtained are not disclosed.

Example No.	Solvent	Yield (%)	Remarks
1	MEK	110.7	
2	MEK	107.0	
3	MEK	106.5	
4	MEK	107.0	
5	MIBK	101.7	
6	THF	95.0	Low yield because of miscibility with water

Calculation of Yield (Example 1):

 $Yield(\%) = (Amount of Resin Solution) \times (solid content) \times 100/ ((Amount of Resorcinol) + (Amount of Formalin))$

$$= 263 \times 0.50 \times 100/(111 + 7.84) = 110.7$$
 (%)

wherein the "Amount of Formalin" =

((Charged Amount) × (solid content (37%))/30 × dehydration (30-18) = (53 ×

•

0.37)/30 × 12 = 7.84

*Molecular weight of formalin: 30

*Molecular weight of water: 18

For all the above reasons, it is respectfully requested that this rejection be REVERSED.

VIII. CONCLUSION

For the above reasons, it is respectfully requested that all the rejections still pending in the Final Office Action be REVERSED.

Customer Number

22850

Tel: (703) 413-3000 Fax: (703) 413 -2220 (OSMMN 06/04)

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Respectfully submitted,

OBLON, SPIVAK, McCLELLAND,

MAIER & NEUSTADT, P.C.

Norman F. Oblon

Harris A. Pitlick

Registration No. 38,779

CLAIMS APPENDIX

- Claim 1. A process for producing a resorcinol-formalin resin containing no inorganic salts, which comprises:
- (1) adding, to a water solvent, resorcinol (A) in an amount of 20 to 150 parts by weight relative to 100 parts by weight of water, an inorganic salt (B) in an amount of 20 to 80 parts by weight relative to 100 parts by weight of water, and an organic solvent (C) having a solubility parameter of 7.0 to 12.5 and capable of dissolving the resorcinol-formalin resin in an amount of 10 to 200 parts by weight relative to 100 parts by weight of resorcinol (A), thereby forming a mixture;
- (2) stirring the mixture at a liquid temperature not higher than the boiling point of the organic solvent (C) to give a two-phase system containing no remaining solid matter;
- (3) adding a catalytic amount of an organic acid or inorganic acid (D) to the mixture formed in step (2);
- (4) adding 1 to 40% formalin (E) dropwise to the mixture formed in step (3) in a molar ratio of formaldehyde/resorcinol of 0.3 to 0.8 under stirring over a period of 1 to 300 minutes thereby forming a reaction system, while maintaining the reaction system at 0 to 60°C;
- (5) stirring the mixture formed in step (4) for further 10 to 60 minutes after the completion of the dropwise addition to cause a liquid-liquid heterogeneous reaction to proceed;
- (6) allowing the reaction system to stand while maintaining it at the temperature of the reaction to separate it into two layers, which are an aqueous layer and a reaction product layer;
 - (7) removing the aqueous layer;

- (8) adding an organic solvent (C) in an amount of 1 to 5 equivalents to the amount of the reaction product to the reaction product layer which is an organic solvent layer to effect dilution;
- (9) adding water to the reaction product layer in an amount which is half of the amount of the organic solvent;
- (10) stirring the reaction system after adding of said organic solvent (C) and said water while maintaining its temperature to be not higher than the boiling point;
- (11) separating the reaction system of step (10) into two layers, including an aqueous layer, after allowing the reaction system to stand; and then
 - (12) removing the aqueous layer to obtain a resorcinol-formalin resin,

wherein said reaction is a one-stage reaction and liquid-liquid distribution is conducted in the same reactor, and

wherein the organic solvent (C) is an organic solvent represented by the following general formula [1]:

General formula [1]:

wherein m represents 0 or 1, n represents 0 or 1, and R₁, R₂, R₃, R₄, and R₅ each independently represents a methyl group, an ethyl group, an n-propyl group, a 2-propyl

group, an n-butyl group, a secondary butyl group, an isobutyl group, or a tertiary butyl group; R_1 and R_2 may be combined to form a ring when m=n=0, R_2 and R_3 may be combined to form a ring when m=1 and n=0, and R_3 and R_4 may be combined to form a ring when m=n=1, with the proviso that when X is -O- and m=n=0, R_1 and R_2 are combined to form a ring.

Claim 2. The process according to claim 1, wherein the 1 to 40% formalin (E) is intermittently added dropwise in a molar ratio of formaldehyde/resorcinol of 0.3 to 0.8 under stirring over a period of 20 to 300 minutes in step (4).

Claim 3. The process according to any one of claims 1 to 2, wherein a peak area corresponding to resorcinol pentanuclear or higher nuclear bodies present in said resorcinol-formalin resin is from 30% to 55% relative to the whole peak area and a peak area corresponding to the resorcinol monomer is from 3% to 9% relative to the whole peak area, the peak areas being obtained by gel permeation chromatographic analysis of the resorcinol-formalin resin.

Claim 4. The process according to claim 1, wherein the organic solvent (C) is an organic solvent having a solubility parameter of 9.0 to 11.0.

Claim 6. The process according to claim 1, wherein the organic solvent (C) is used as a mixture of two or more thereof.

Claim 7. The process according to claim 1, wherein the inorganic salt (B) is a salt formed from one or two or more cations selected from alkali metals and alkaline earth metals

and one or two or more anions selected from a sulfate ion, a nitrate ion, a chlorine ion, a bromine ion, an iodine ion, and a thiocyanate ion.

Claim 8. The process according to claim 1, wherein the inorganic salt (B) is calcium chloride.

Claim 9. The process according to claim 1, wherein the amount of the organic solvent (C) added in step (1) is from 30 to 100 parts by weight relative to 100 parts by weight of resorcinol (A).

Claim 10. The process according to claim 1, wherein the organic acid or inorganic acid (D) is hydrochloric acid.

Claim 11. The process according to claim 1, wherein the mole number of formaldehyde in the formalin (E) relative to the mole number of resorcinol (A) is in a molar ratio of formaldehyde/resorcinol of 0.5 to 0.8.

Claim 12. The process according to claim 1, wherein time for the dropwise addition of the formalin (E) is from 20 to 120 minutes.

Claim 13. The process according to claim 1, wherein after step (12), water is added in an amount of 1 to 10 equivalents by weight to the resorcinol-formalin resin in the organic solvent (C) solution of the resorcinol-formalin resin and the organic solvent (C) is removed by distillation to finally obtain an aqueous resorcinol-formalin resin solution having a reaction product concentration of 30 to 80%.

Claim 14. The process according to claim 1, wherein after step (12), the organic solvent (C) is added to the organic solvent layer obtained by the separation into two layers after allowing to stand and the removal of the aqueous layer, in an amount of 2 to 10 equivalents to the weight of the reaction product to effect dilution, water is removed by distillation at the azeotropic temperature of water and the organic solvent, and then solid matter is removed by filtration after cooling to room temperature.

- Claim 15. A resorcinol-formalin resin containing no inorganic salts, produced by a production process comprising:
- (1) adding, to a water solvent, resorcinol (A) in an amount of 20 to 150 parts by weight relative to 100 parts by weight of water, an inorganic salt (B) in an amount of 20 to 80 parts by weight relative to 100 parts by weight of water, and an organic solvent (C) having a solubility parameter of 7.0 to 12.5 and capable of dissolving the resorcinol-formalin resin in an amount of 10 to 200 parts by weight relative to 100 parts by weight of resorcinol (A), thereby forming a mixture;
- (2) stirring the mixture at a liquid temperature not higher than the boiling point of the organic solvent (C) to give a two-phase system containing no remaining solid matter;
- (3) adding a catalytic amount of an organic acid or inorganic acid (D) to the mixture formed in step (2);
- (4) adding 1 to 40% formalin (E) dropwise to the mixture formed in step (3) in a molar ratio of formaldehyde/resorcinol of 0.3 to 0.8 under stirring over a period of 1 to 300 minutes thereby forming a reaction system, while maintaining the reaction system at 0 to 60°C:

- (5) stirring the mixture formed in step (4) for further 10 to 60 minutes after the completion of the dropwise addition to cause a liquid-liquid heterogeneous reaction to proceed;
- (6) allowing the reaction system to stand while maintaining it at the temperature of the reaction to separate it into two layers, which are an aqueous layer and a reaction product layer;
 - (7) removing the aqueous layer;
- (8) adding an organic solvent (C) in an amount of 1 to 5 equivalents to the amount of the reaction product to the reaction product layer which is an organic solvent layer to effect dilution;
- (9) adding water to the reaction product layer in an amount which is half of the amount of the organic solvent;
- (10) stirring the reaction system after adding of said organic solvent (C) and said water while maintaining its temperature to be not higher than the boiling point;
- (11) separating the reaction system of step (10) into two layers, including an aqueous layer, after allowing the reaction system to stand; and then
 - (12) removing the aqueous layer to obtain a resorcinol-formalin resin,

wherein said reaction is a one-stage reaction and liquid-liquid distribution is conducted in the same reactor, and

wherein the organic solvent (C) is a ketone represented by the following general formula [1]:

General formula [1]:

$$\left(R_3-Y \xrightarrow{}_m R_1-X-R_2 \xrightarrow{} \left(Z-R_4\right)_n\right)$$

wherein m represents 0 or 1, n represents 0 or 1, and R_1 , R_2 , R_3 , R_4 , and R_5 each independently represents a methyl group, an ethyl group, an n-propyl group, a 2-propyl group, an n-butyl group, a secondary butyl group, an isobutyl group, or a tertiary butyl group; R_1 and R_2 may be combined to form a ring when m=n=0, R_2 and R_3 may be combined to form a ring when m=1 and n=0, and R_3 and R_4 may be combined to form a ring when m=n=1.

Claim 16. The resorcinol-formalin resin according to claim 15, wherein the 1 to 40% formalin (E) is intermittently added dropwise in a molar ratio of formaldehyde/resorcinol of 0.3 to 0.8 under stirring over a period of 20 to 300 minutes in step (4).

Claim 17. The resorcinol-formalin resin according to any one of claims 15 to 16, wherein a peak area corresponding to resorcinol pentanuclear or higher nuclear bodies present in said resorcinol-formalin resin is from 30% to 55% relative to the whole peak area and a peak area corresponding to the resorcinol monomer is from 3% to 9% relative to the

whole peak area, the peak areas being obtained by gel permeation chromatographic analysis of the resorcinol-formalin resin.

Claim 18. The resorcinol-formalin resin according to claim 15, wherein the organic solvent (C) is an organic solvent having a solubility parameter of 9.0 to 11.0.

Claim 20. The resorcinol-formalin resin according to claim 15, wherein the organic solvent (C) is used as a mixture of two or more thereof.

Claim 21. The resorcinol-formalin resin according to claim 15, wherein after step (12), water is added in an amount of 1 to 10 equivalents by weight to the resorcinol-formalin resin in the organic solvent (C) solution of the resorcinol-formalin resin and the organic solvent (C) is removed by distillation to finally obtain an aqueous resorcinol-formalin resin solution having a reaction product concentration of 30 to 80%.

Claim 22. The resorcinol-formalin resin according to claim 15, wherein after step (12), the organic solvent (C) is added to the organic solvent layer obtained by the separation into two layers after allowing to stand and the removal of the aqueous layer, in an amount of 2 to 10 equivalents to the weight of the reaction product to effect dilution, water is removed by distillation at the azeotropic temperature of water and the organic solvent, and then solid matter is removed by filtration after cooling to room temperature.

Claim 23 (New): The resorcinol-formalin resin according to claim 15, wherein the organic solvent (C) is methyl ethyl ketone or methyl isobutyl ketone.

EVIDENCE APPENDIX

None.

RELATED PROCEEDINGS APPENDIX

None.